

Thermal and dielectric investigation on the effect of UV radiation on LR-115 detector

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Abstract : Non-isothermal studies were carried out using thermogravimetry (TG) and differential thermogravimetry (DTG) measurements to obtain the thermal activation energy of decomposition for LR-115 solid state nuclear track detector before and after exposure to different UV energy fluences ranging from 11.3 to 113 kJ/cm². Thermal gravitational analysis (TGA) indicated that both unirradiated and irradiated LR-115 samples decomposed in one main breakdown stage. Kinetic analysis has been carried out to determine the thermal activation energy of decomposition, a decrease in activation energies was observed on increasing the exposure to UV. Also, the variation of the transition temperatures with the UV energy fluence have been determined using differential scanning calorimeter (DSC). In addition, dielectric measurements have been made in the frequency range 100 Hz–100 kHz. The measurements were carried out in the temperature range 25–90°C. The dielectric constant ϵ' and dielectric loss ϵ'' showed an increase on increasing the temperature up to 90°C. Also the frequency dependence of ϵ' is characterized by a primary relaxation process. Moreover, the effect of UV radiation on ϵ' and ϵ'' have been measured at room temperature 25°C and at 100 kHz. The results indicated that both ϵ' and ϵ'' showed an increase on increasing the UV energy fluence due to the degradation caused by the UV light.

Keywords : UV radiation, Thermal properties, Dielectric properties and LR-115 detector

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1. Introduction

Although the common cellulose derivatives are among the most investigated of all high polymers, we have been prompted to undertake another study for several reasons. First, cellulose derivatives appear to be almost the only unbranched polymers readily available in a broad range of molecular weights. Secondly, the study of the thermal properties of

polymers is important to check their thermal stability and thus their suitability for service. Third, due to their potential application in high-energy density solid-state batteries, the temperature and frequency dependencies of the dielectric properties of polymers have been studied.

Numerous publications have dealt with the temperature and frequency dependencies of the dielectric properties of polymers [1–5]. Also the effect of UV radiation on the physical and optical properties of polymers has been studied [6,7]. Nouh *et al.* [8] studied the effect of non ionizing radiations on the degree of ordering of LR-115 detector and on the thermal properties of some polymers.

This paper deals with the investigation of the effect of UV radiation on the thermal and dielectric properties of LR-115 polymer aiming to (1) know the mechanism of thermal degradation of LR-115 polymer to be able to choose the ways of stabilizing it and, consequently, of prolonging the service life of articles made from LR-115 polymer, (2) obtain optimum dielectric insulating characteristics for engineering applications, (3) introduce the basis which may be used in constructing a simple sensor for irradiation.

2. Experimental

A) Samples :

LR-115 sheets used in this study were manufactured by Kodak Pathe (France). It consists of a sensitive cellulose nitrate layer of 12 μm thickness on a 100 μm thick polyester support and its density is 1.42 gm/cm^3 .

B) Irradiation facilities :

All samples were exposed to different UV energy fluences using a Hg lamp of 50 W, at 50/60 Hz, with an illumination flux of 3.14 W/cm^2 . All the measurements were carried out 24 h after UV exposure.

C) Experimental apparatus :

The thermal behavior was investigated using differential scanning calorimeter (DSC) and thermal gravitational analysis (TGA) type Shimadzu-50. $\alpha\text{-Al}_2\text{O}_3$ powder was used as a reference for DSC measurements. Thermal experiments were carried out on all samples at different heating rates (5, 10, 15 and 20°C/min) with N_2 as a carrier gas at a flow rate of 30 cm^3/min . The values obtained for transition temperatures were measured with a basic accuracy of $\pm 0.1^\circ\text{C}$.

All the dielectric measurements were carried out using RCL bridge Type PM6304/031 programmable automatic RCL meter measures in the range from 50Hz to 100 kHz. The temperatures were controlled using an ultrathermostate. The capacitance was measured with a basic accuracy of 0.2 to 0.3%. Also, a capacitance accuracy test was made by connecting a stable capacitor to the instrument and reading the display to verify that the

3. Results and Discussion

1. Thermal properties :

A) Transition temperatures :

Thermal characterization gives a fairly good idea about the changes taking place in polymers. The DSC thermograms measured in the temperature range from room temperature up to 320°C for unirradiated and irradiated LR-115 samples are shown in Figure 1. All the thermograms were characterized by the appearance of two peaks, one endothermic peak due to the melting temperature T_m and one exothermic peak due to the

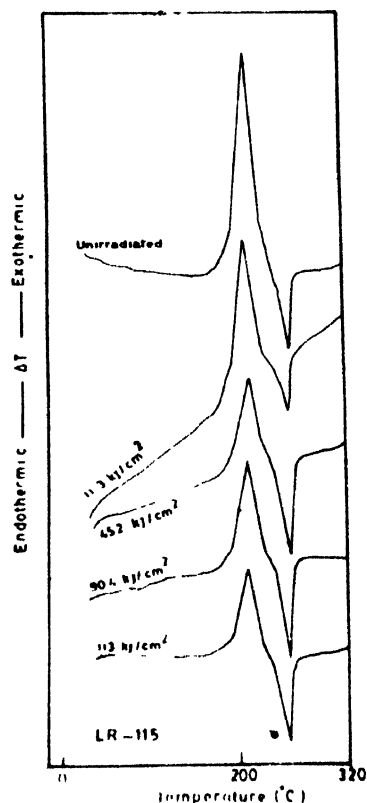


Figure 1. DSC thermograms measured in the temperature range from room temperature up to 320°C for unirradiated and irradiated LR-115 samples

crystallization temperature T_c . The glass transition temperature could be detected for some samples. The values obtained for these transition temperatures are given in Table 1. The values indicated that both the crystallization and melting temperatures exhibited the same trends with the radiation dose, where, they showed an increase in magnitude up to a maximum value around 45.2 kJ/cm² of irradiation followed by a decrease on increasing the UV energy fluence due to the thermal degradation caused by the UV light.

Table 1. Transition temperatures and values of thermal activation energy of decomposition for LR-115 (the values obtained for T_g , T_c and T_m are $\pm 0.1^\circ\text{C}$).

UV energy fluence (kJ/cm^2)	$T_g(^{\circ}\text{C})$	$T_c(^{\circ}\text{C})$	$T_m(^{\circ}\text{C})$	$E_a(\text{eV})$
0.0	88.2	208.8	256.8	3.66
11.3	—	209.0	257.0	3.48
45.2	—	210.0	257.2	3.41
90.4	90.1	209.8	256.9	3.20
113.0	90.2	208.9	255.6	2.98

B) Activation energy of decomposition :

Evaluation of the activation energy of decomposition is useful for studying the thermal stability of materials. Thermal gravitational analysis (TGA) provides quantitative information on weight change process while differential thermal gravitational (DTG) provides rate of weight loss (dW/dT). TGA measurements were performed on all irradiated and unirradiated LR-115 samples at different heating rates (5, 10, 15 and $20^\circ\text{C}/\text{min}$). Figure 2 shows TGA and DTG thermogram for unirradiated LR-115 sample as an example.

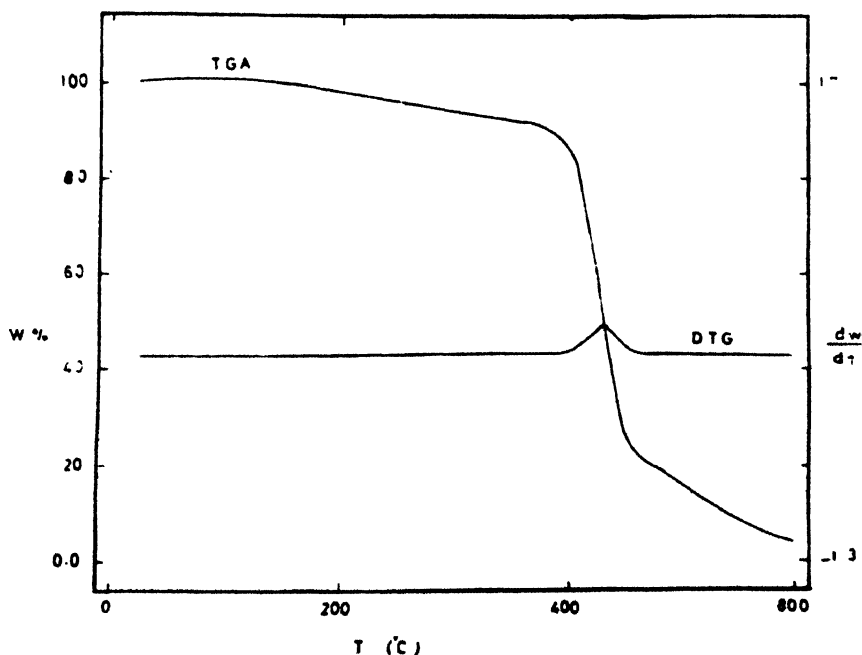


Figure 2. TGA and DTG thermogram for unirradiated LR-115 sample (as an example).

It is clear that LR-115 detector decomposes in one weight loss stage. Various thermogravimetric methods have been proposed for the measurement of activation energies.

The method proposed by Ozawa [9] has been used in the present work. In this method, TG curves obtained at different heating rates are required where the following equation is valid :

$$\log \Phi_1 + 0.4567 E/RT_1 = \log \Phi_2 + 0.4567 E/RT_2 = \dots$$

where T_i is the temperature at which the sample is decomposed at a heating rate Φ_i .

According to the above equation, a plot of the logarithms of the heating rates against the reciprocal of absolute temperature, leads to a straight line relationship in the range where the decomposed ratios are equal. Hence, the activation energy of decomposition E_a can be evaluated from the slope of the line. The values of thermal activation energy of decomposition E_a were calculated for all irradiated and unirradiated LR-115 samples using TGA and DTG curves and are given in Table 1. The values obtained indicates that E_a decreases in magnitude on increasing the UV energy fluence. This may be attributed to the thermal degradation, where, under prolonged heating, there occurring a random breaking of bonds and sometimes the detachment of low-molecular products because of reactions of side groups without any appreciable change in the initial molecular weight.

2. Dielectric properties :

A) The frequency dependence of the dielectric constant and dielectric loss :

The frequency (f) dependence of the dielectric constant ϵ' of unirradiated and irradiated LR-115 samples at room temperature (25°C) is shown in Figure 3. The graph shows that ϵ'

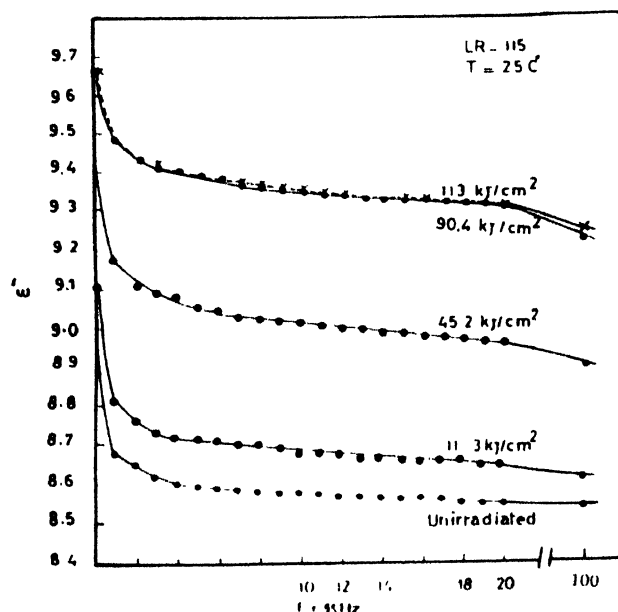


Figure 3. The dependence of dielectric constant (ϵ') measured at a temperature of 25°C on the frequency for unirradiated and irradiated LR-115 samples.

decreases almost linearly with frequency (f) in the frequency range from 4 to 20 kHz. The strong low-frequency dispersion that characterizes the frequency dependence of the

dielectric constant ϵ' (Figure 3) is to be ascribed to a dipole-orientation relaxation process [10]. This process normally involves a rapid increase in dielectric constant as the measurement frequency is reduced. The variation of dielectric loss ϵ'' with frequency for all the unirradiated and irradiated LR-115 samples is shown in Figure 4. From the figure it

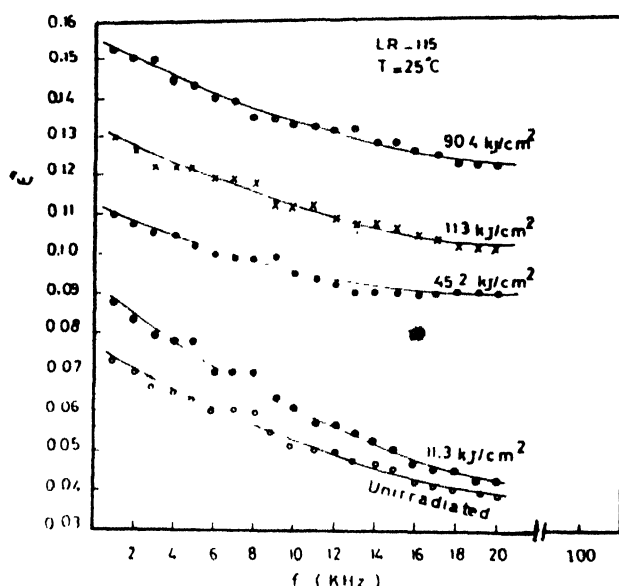


Figure 4. The dependence of dielectric loss (ϵ'') measured at a temperature of 25°C on the frequency for unirradiated and irradiated LR-115 samples

is clear that ϵ'' decreases almost linearly in the frequency range 4–20 kHz. While, on increasing the frequency up to 100 kHz, a large drop in ϵ'' values was observed. For this reason, the values of ϵ'' at 100 kHz for all the LR-115 samples are given in Table 2 and

Table 2. Values of ϵ' and ϵ'' measured at 100 kHz and 25°C for LR-115 (the values obtained for ϵ' and ϵ'' are ± 0.2 to 0.3%).

UV energy fluence (kJ/cm ²)	ϵ'	ϵ''
0.0	8.53	0.009
11.3	8.61	0.017
45.2	8.90	0.045
90.4	9.23	0.065
113.0	9.25	0.065

only the straight portion (4–20 kHz) is shown in Figure 4. The dielectric loss relaxation obtained in Figure 4 is almost due to the micro-Brownian motions of the chain (glass-rubber relaxation) in the non crystalline regions of the polymer [11].

B) Temperature dependence of the dielectric constant and dielectric loss :

The temperature dependencies of the dielectric constant and dielectric loss of unirradiated LR-115 sample at 100 kHz are given in Figure 5. From the figure it is clear that the rate of increase of ϵ' and ϵ'' with temperature is slow since it depends on the degree of polymer

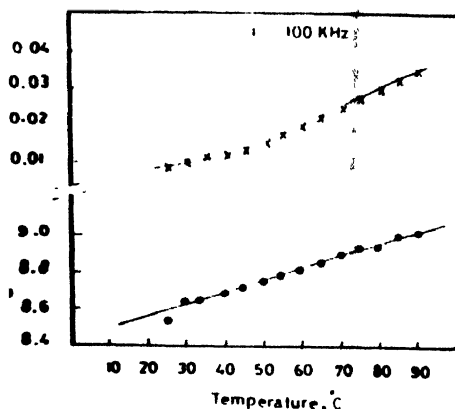


Figure 5. The dependence of dielectric constant (ϵ') and dielectric loss (ϵ'') measured at 100 kHz on the temperature for unirradiated LR-115 detector.

hardness. The higher the hardness the higher the rate of increase, as the temperature causes loosening up of the rigid structure, resulting in an increase in dipole orientation and hence an increase in ϵ' . These results show that LR-115 plastic detector possess better dielectric properties at temperatures higher than room temperature and this property is useful for electrical insulating purposes. Also the interpretation of Figure 5 can be given as follows :

At temperatures above the glass transition temperature, T_g , the motion of long-chain molecules occurs as a result of the cooperative movement of segments of the chain. The rate of segmental motion can be estimated from the relaxation time of the dielectric relaxation process associated with the micro-brownian motion of the main chain. This rate can be thought of as a function of the free volume fraction which varies with temperature.

C) The effect of UV energy fluence on the dielectric constant and dielectric loss :

The UV energy fluence dependencies of the dielectric constant and dielectric loss for LR-115 detector at 100 kHz and 25°C are given in Table 2. The values obtained indicated that both ϵ' and ϵ'' showed almost an increase on increasing the exposure to UV. This increase may be due to degradation, where at degradation, the standard chains and greater number of chain ends lead to weakening and often to embrittlement, even the material may have become somewhat softer. Thus the increase in ϵ' with the physical degradation is related to the increase in the degree of soft segments and the loss in hard segments.

4. Conclusions

From the above study one can draw the following conclusions :

- (1) Exposing the LR-115 polymer to UV causes a random breaking of bonds which results in a decrease in thermal activation energy of decomposition. The TGA measurements indicated that when LR-115 polymer is heated, its thermal stability is usually appraised from the loss of mass and its molecular weight sharply diminishes which is due to the degradation of the molecular chains.
- (2) The values of the dielectric constant and dielectric loss of LR-115 detector are dependent on the frequency, temperature and UV energy fluence.
- (3) LR-115 detector was found to possess better insulating properties at temperatures higher than room temperature and thus it can be used for electrical insulating purposes.

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